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March 29, 2002

BOX PCT

Commissioner for Patents
Washington, D.C. 20231

PCT/JP00/06631
-filed September 27, 2000

Re: Application of Sadashige IRIE, Kazuyoshi MIMURA,
Hirofumi NISHIBAYASHI, Hiroyuki TANAKA, Tsuyoshi NOGUCHI, and
Mitsuru KISHINE
TRANSPARENT ELASTOMER COMPOSITION
Assignee: DAIKIN INDUSTRIES, LTD.
Our Ref: Q69144

Dear Sir:

The following documents and fees are submitted herewith in connection with the above application for the purpose of entering the National stage under 35 U.S.C. § 371 and in accordance with Chapter II of the Patent Cooperation Treaty:

- ☒ an executed Declaration and Power of Attorney.
- ☒ an English translation of the International Application.
- ☒ two (2) sheets of drawings.
- ☒ an executed Assignment and PTO 1595 form.
- ☒ Notification Concerning Submission or Transmittal or Priority Document
- ☒ International Search Report, Information Disclosure Statement, PTO Form 1449 with references.
- ☒ a Preliminary Amendment

It is assumed that copies of the International Application, the International Search Report, the International Preliminary Examination Report, and any Articles 19 and 34 amendments as required by § 371(c) will be supplied directly by the International Bureau, but if further copies are needed, the undersigned can easily provide them upon request.

The Government filing fee, after entry of Preliminary Amendment, is calculated as follows:

Total claims	16	-	20	=		x	\$18.00	=	\$0.00
Independent claims	2	-	3	=		x	\$84.00	=	\$0.00
Base Fee									\$890.00

TOTAL FILING FEE	\$890.00
Recordation of Assignment	\$ 40.00
TOTAL FEE	<u>\$930.00</u>



SUGHRUE MION, PLLC

National Stage Entry of PCT/JP00/06631

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Checks for the statutory filing fee of \$890.00 and Assignment recordation fee of \$40.00 are attached. You are also directed and authorized to charge or credit any difference or overpayment to Deposit Account No. 19-4880. The Commissioner is hereby authorized to charge any fees under 37 C.F.R. §§ 1.16, 1.17 and 1.492 which may be required during the entire pendency of the application to Deposit Account No. 19-4880. A duplicate copy of this transmittal letter is attached.

Priority is claimed from:

<u>Country</u>	<u>Application No</u>	<u>Filing Date</u>
Japan	277995/1999	September 30, 1999

Respectfully submitted,

A handwritten signature in black ink, appearing to read 'A. Rosner'.

Abraham J. Rosner
Registration No. 33,276

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Date: March 29, 2002

10/089523

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PATENT APPLICATION

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of

Sadashige IRIE, et al.

Appln. No.: National Stage Entry of PCT/JP00/06631

Confirmation No.: NOT YET ASSIGNED

Group Art Unit: NOT YET ASSIGNED

Filed: March 29, 2002

Examiner: NOT YET ASSIGNED

For: TRANSPARENT ELASTOMER COMPOSITION

PRELIMINARY AMENDMENT

Commissioner for Patents
Washington, D.C. 20231

Sir:

Prior to examination, please amend the above-identified application as follows:

IN THE CLAIMS:

Please enter the following amended claims:

3. (Amended) The composition of Claim 1, wherein the elastomer is a fluorine-containing elastomer.
4. (Amended) The composition of Claim 1, wherein the composition contains a crosslinking agent.
5. (Amended) The composition of Claim 1, wherein the transparent elastomer composition has a haze value of not more than 50%.
8. (Amended) An elastomeric molded article which is obtained by vulcanizing and molding the composition of Claim 1.

PRELIMINARY AMENDMENT
National Stage Entry of PCT/JP00/06631

Please add the following new claims:

11. (New) The composition of Claim 2, wherein the elastomer is a fluorine-containing elastomer.
12. (New) The composition of Claim 2, wherein the composition contains a crosslinking agent.
13. (New) The composition of Claim 2, wherein the transparent elastomer composition has a haze value of not more than 50%.
14. (New) An elastomeric molded article which is obtained by vulcanizing and molding the composition of Claim 2.
15. (New) The elastomeric molded article of Claim 14, which is transparent.
16. (New) The elastomeric molded article of Claim 15, which has a haze value of not more than 50%.

PRELIMINARY AMENDMENT
National Stage Entry of PCT/JP00/06631

REMARKS

Entry and consideration of this Amendment is respectfully requested.

Respectfully submitted,



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Date: March 29, 2002

APPENDIX

VERSION WITH MARKINGS TO SHOW CHANGES MADE

IN THE CLAIMS:

The claims are amended as follows:

3. (Amended) The composition of Claim 1, ~~or 2~~ wherein the elastomer is a fluorine-containing elastomer.

4. (Amended) The composition of ~~any of Claims~~ Claim 1, ~~to 3~~ wherein the composition contains a crosslinking agent.

5. (Amended) The composition of ~~any of Claims~~ Claim 1, ~~to 4~~ wherein the transparent elastomer composition has a haze value of not more than 50%.

8. (Amended) An elastomeric molded article which is obtained by vulcanizing and molding the composition of ~~any of Claims~~ Claim 1 ~~to 5~~.

Claims 11 through 16 are added as new claims.

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JC13 Rec'd PST/PTC 29 MAR 2002

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DESCRIPTION

TRANSPARENT ELASTOMER COMPOSITION

5

TECHNICAL FIELD

The present invention relates to a transparent composition comprising an elastomer and fine particles of fluorine-containing resin, for example, a transparent elastomer composition comprising a fluorine-containing elastomer as a matrix and fine particles of fluorine-
10 containing resin which are finely dispersed in the elastomer. The elastomer composition can provide a molded article possessing enhanced mechanical strength and is useful as a base elastomer for various molded articles. Further the elastomer composition can provide an elastomeric molded article which is excellent in mechanical
15 strength and transparency by selecting an additive to be added thereto.

BACKGROUND ART

It has been known that a fluorine-containing resin is added to a fluorine-containing elastomer. An object of the addition is to
20 enhance a mechanical strength by using the fluorine-containing resin as a clean filler and to utilize a friction property thereof. Also, for mixing the resin and the elastomer, there are proposed a dry blend method by using a usual rubber mixing roll, a method for enhancing a dispersibility by using a solvent, and the like.

25 For example, in JP-A-55-151051, a fluorine-containing rubber excellent in abrasion resistance is obtained by blending a low molecular weight polytetrafluoroethylene (PTFE). Also, JP-A-63-

178149 describes a method of adding and mixing a PTFE powder to a fluorine-containing rubber solution in order to enhance a gas barrier property and mechanical strength of a rubber. Further, JP-A-2-261850 proposes that a fluorine-containing resin is blended in a large
5 amount of from 30 to 150 parts by weight together with an organic solvent to 100 parts by weight of a rubber to lower a friction coefficient and enhance mechanical properties.

Example of an addition of a fluorine-containing resin for obtaining cleanliness is described in WO97/08239 which discloses that
10 a composition prepared by adding 5 to 50 parts by weight of a fluorine-containing resin powder having an average particle size of from 0.2 to 50 μm to 100 parts by weight of a fluorine-containing rubber is excellent as a sealing agent for etching equipment. Also, WO95/02634 discloses that a composition prepared by adding 2 to 50 parts by weight of a
15 fluorine-containing resin fine powder to 100 parts by weight of a rubber component is a clean composition suitable for a wet process of semiconductors.

However those prior techniques are directed to addition and mixing of a fluorine-containing resin to a matrix rubber (elastomer), and
20 even if a dispersibility is enhanced by using a solvent, as a particle size becomes finer, a uniform dispersion is difficult to obtain. This is shown by a fact that for example, when fine particles of a fluorine-containing resin are dispersed in a transparent elastomer, if they are dispersed uniformly, the composition should be transparent, but actually a
25 transparent elastomer composition in which a fluorine-containing resin is finely uniformly dispersed has not yet been obtained.

If fine particles of a fluorine-containing resin are not

dispersed uniformly, a strength of a molded article obtained by vulcanizing and molding is low and an effect of adding the fluorine-containing resin is not obtained sufficiently.

It is therefore an object of the present invention is to provide
5 an elastomer composition which is in a state that fluorine-containing resin fine particles are dispersed uniformly in a transparent elastomer, and can exhibit the maximum of effect of adding the fluorine-containing resin.

10 DISCLOSURE OF INVENTION

Namely, the present invention relates to a transparent elastomer composition in which fine particles of a fluorine-containing resin are finely dispersed in a transparent elastomer.

It is preferable that the transparent elastomer as a matrix is a
15 fluorine-containing elastomer. Also it is preferable that the fine particles of a fluorine-containing resin have an average particle size of from 20 to 150 nm.

The present invention also relates to an elastomer composition containing a crosslinking agent and as the case demands, a
20 crosslinking accelerator and a filler.

The elastomer composition of the present invention can be prepared, for example, by mixing an emulsion of transparent elastomer particles and an emulsion of fluorine-containing resin fine particles and then co-coagulating the mixture.

25 The present invention also relates to an elastomeric molded article obtained by vulcanizing and molding the above-mentioned elastomer composition, and further relates to a transparent elastomeric

molded article.

In the present invention, being transparent means that a haze value mentioned hereinbelow is not more than 50 %.

5

BRIEF DESCRIPTION OF DRAWINGS

Fig. 1 is a photograph ($\times 20000$) taken by a transmission type electron microscope of the elastomer composition obtained in Example 1 of the present invention.

Fig. 2 is a photograph ($\times 20000$) taken by a transmission type electron microscope of the elastomer composition obtained in Comparative Example 3.

BEST MODE FOR CARRYING OUT THE INVENTION

In the present invention, uniform dispersion of fine particles of a fluorine-containing resin which has been difficult to obtain is achieved by the method of mixing an emulsion of transparent elastomer particles and an emulsion of fluorine-containing resin fine particles and then co-coagulating the mixture.

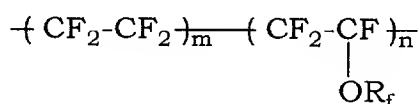
Whether or not the fluorine-containing resin fine particles are dispersed finely uniformly can be seen obviously by observing through a transmission type electron microscope (TEM) (cf. Fig. 1 explained hereinbelow) or can be judged by determining whether or not transparency of the transparent matrix elastomer is maintained even after mixing of the fluorine-containing resin fine particles. In other words, the mixture obtained by dispersing is turbid in white even if the dispersing is carried out by a dry blend method in which the mixing is carried out after the fluorine-containing resin fine particles having the

same fine particle size are once coagulated and dried or by a method of mixing by using a solvent. It can be considered that this is because the fluorine-containing resin fine particles are subject to secondary agglomeration or are dispersed locally (cf. Fig. 2 explained hereinbelow).

5 The elastomer composition of the present invention before crosslinking has a transparency of not more than 50 %, preferably not more than 40 %, particularly not more than 30 % in a haze value.

 The matrix elastomer to be used in the present invention is good as far as it is a transparent elastomer capable of forming an
10 emulsion in the form of particles and has affinity for the fluorine-containing resin. From this point of view, a fluorine-containing elastomer is preferable.

 Examples of the fluorine-containing elastomer are, for instance,
15 copolymer elastomers (tetrafluoroethylene (TFE)/perfluoro(alkyl vinyl ether) (PAVE) elastomers) represented by the formula (1):

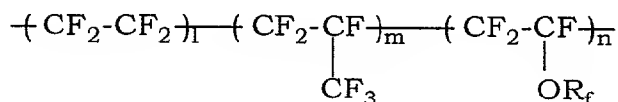


20

wherein $m/n = 95$ to $50/5$ to 50 (% by mole, hereinafter the same), R_f is a perfluoroalkyl group having 1 to 8 carbon atoms,

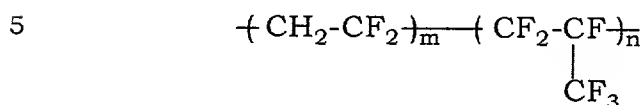
terpolymer elastomers (TFE/hexafluoropropylene (HFP)/PAVE elastomers) represented by the formula (2):

25



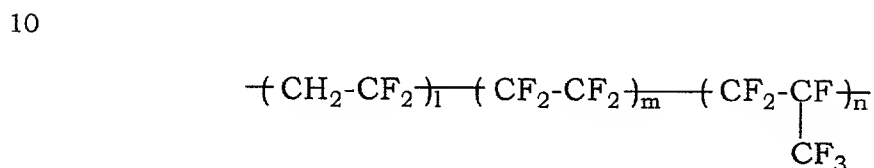
wherein $1/m/n = 95$ to $35/0$ to $30/5$ to 35 , R_f is a perfluoroalkyl group having 1 to 8 carbon atoms,

copolymer elastomers represented by the formula (3):



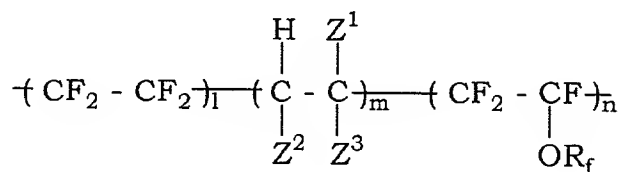
wherein $m/n = 85$ to $60/15$ to 40 ,

terpolymer elastomers represented by the formula (4):



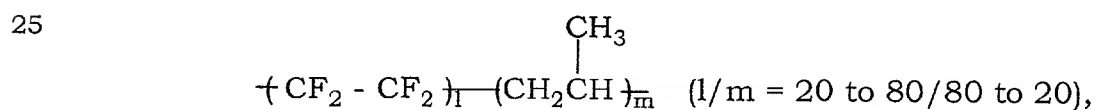
wherein $1/m/n = 85$ to $20/0$ to $40/15$ to 40 ,

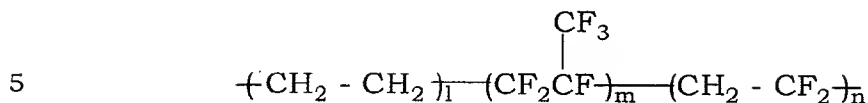
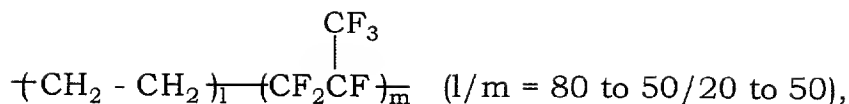
15 terpolymer elastomers represented by the formula (5):



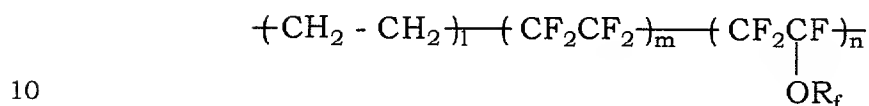
20

wherein $1/m/n = 95$ to $45/0$ to $10/5$ to 45 , Z^1 , Z^2 and Z^3 are fluorine atoms or hydrogen atoms independently, R_f is a perfluoroalkyl group having 1 to 8 carbon atoms,





(l/m/n = 10 to 65/15 to 45/0.1 to 45),



(l/m/n = 1 to 80/0 to 80/10 to 50, R_f is as defined above),

and the like.

15 More concretely there are TFE/PAVE copolymer elastomer, vinylidene fluoride (VdF)/hexafluoropropylene (HFP) copolymer elastomer, VdF/HFP/TFE copolymer elastomer, VdF/TFE/PAVE copolymer elastomer, and the like. A small amount of monomer having a crosslinkable reaction group may be copolymerized with those
20 elastomers. Examples of the crosslinkable reaction group are, for instance, iodine atom, bromine atom, nitrile group, carboxyl group, unsaturated double bond, hydroxyl group, and the like.

Those fluorine-containing elastomers can be prepared by usual emulsion polymerization method, and the obtained polymerization
25 reaction product which is an emulsion can be used as it is for co-coagulation to be described hereinbelow, or can be used for the co-coagulation after optionally adjusting a concentration thereof. The

emulsion may be once dried and then formed into an emulsified dispersion again.

Examples of the elastomer other than the fluorine-containing elastomer are, for instance, hydrogenated nitrile butadiene rubber, acrylic rubber, silicone rubber, and the like.

An average particle size of the elastomer particles in the emulsion is not limited particularly, and is, for example, from 10 to 800 nm, preferably from 20 to 500 nm. However when the average particle size is less than 10 nm, the coagulation becomes difficult, and when the average particle size is more than 800 nm, the emulsion becomes unstable and the co-coagulation becomes difficult.

The fine particles of the fluorine-containing resin to be finely dispersed into the elastomer are not limited particularly. For example, there are fine particles of:

- (1) polytetrafluoroethylene (PTFE);
- (2) TFE/ $\text{CF}_2=\text{CF}-\text{O}-\text{R}_f^1$ (FVE) copolymer (Proportion thereof is one exhibiting a non-elastomeric property, for example, a proportion of $\text{CF}_2=\text{CF}-\text{O}-\text{R}_f^1$ is not more than 15 % by mole. R_f^1 is a linear or branched fluoro- or perfluoroalkyl group or a fluoro- or perfluorooxyalkyl group which may have an ether type oxygen atom.). For example, TFE/PAVE copolymer (PFA);
- (3) TFE/ $\text{CF}_2=\text{CF}-\text{R}_f^1$ copolymer (Proportion thereof is one exhibiting a non-elastomeric property, for example, a proportion of $\text{CF}_2=\text{CF}-\text{R}_f^1$ is not more than 15 % by mole. R_f^1 is as defined above.) For example, TFE/HFP copolymer (FEP);
- (4) Ethylene/TFE (30 to 60/70 to 40 in % by mole, hereinafter the same) copolymer;

- (5) Polychlorotrifluoroethylene (PCTFE);
- (6) Ethylene/chlorotrifluoroethylene (CTFE) (30 to 60/70 to 40) copolymer;
- (7) Polyvinylidene fluoride (PVdF);
- 5 (8) Vinylidene fluoride (VdF)/TFE (70 to 99/30 to 1) copolymer;
- (9) VdF/TFE/CTFE (50 to 99/30 to 0/20 to 1) copolymer;
- (10) VdF/TFE/HFP (60 to 99/30 to 0/10 to 1) copolymer;
- (11) Ethylene/TFE/HFP (6 to 60/40 to 81/1 to 30) copolymer;
- (12) 3,3,3-trifluoropropylene-1,2-trifluoromethyl-3,3,3-
- 10 trifluoropropylene-1/PAVE (40 to 60/60 to 40) copolymer;

and the like. Among them, when a low friction property is imparted to the molded article, the above-mentioned (1) is preferred. Particularly in order to enhance compatibility with the perfluoroelastomer component, the above-mentioned (2) and (3) are preferred.

15 The PTFE of the above-mentioned (1) encompasses not only TFE homopolymer but also a modified PTFE obtained by copolymerizing a comonomer in such a small amount as not giving melt-flowability. Examples of the comonomer are HFP, CTFE, perfluorovinylether, trifluoroethylene, perfluoroalkylethylene, and the like. When
20 perfluorovinylether is copolymerized as a comonomer, its amount is up to 2 % by weight, preferably from 0.001 to 1 % by weight, more preferably from 0.01 to 1 % by weight.

Those fluorine-containing resins can be prepared by usual emulsion polymerization method, and the obtained polymerization
25 reaction product which is an emulsion can be used as it is for co-coagulation to be described hereinbelow or can be used for the co-coagulation after optionally adjusting a concentration thereof. The

emulsion may be once dried and then formed into an emulsified dispersion again.

An average particle size of the fluorine-containing resin fine particles in the emulsion is less than 150 nm, preferably from 20 to 150 nm, more preferably from 20 to 100 nm. When the average particle size is too small, productivity is lowered remarkably, and when the average particle size is not less than 200 nm, a uniform dispersion cannot be obtained.

The mixing ratio of the elastomer and the fluorine-containing resin may be selected optionally depending on properties to be imparted to the molded article. It is desirable that the amount of the fluorine-containing resin is not less than 1 part by weight, preferably not less than 5 parts by weight based on 100 parts by weight of the elastomer from the point of obtaining a reinforcing effect, and is not more than 150 parts by weight, preferably not more than 100 parts by weight, more preferably not more than 50 parts by weight based on 100 parts by weight of the elastomer from the viewpoint of easiness in processing of the obtained crosslinked rubber.

The combination of the elastomer with the fluorine-containing resin may be selected to intended functions in consideration of whether or not coagulating property thereof is approximate to each other and whether or not those polymers have affinity.

Non-restricted examples of the preferable combination are, for instance,

- (a) TFE/PAVE elastomer with TFE/PAVE resin (PFA);
- (b) TFE/PAVE elastomer with TFE/HFP resin (FEP);
- (c) VdF/HFP elastomer with PVdF resin;

- (d) VdF/HFP/TFE elastomer with PVdF resin;
 - (e) TFE/propylene elastomer with ethylene/TFE resin (ETFE);
- and the like.

The elastomer composition of the present invention can be prepared by mixing the above-mentioned emulsion of the elastomer particles to the emulsion of the fluorine-containing resin fine particles and then coagulating the mixture. The co-coagulation can be carried out by known coagulation method. For example, there can be used a method of adding a solution of the mixture of the emulsion of the elastomer particles and the emulsion of the fluorine-containing resin fine particles dropwise into a coagulating solution, a method of adding a coagulating solution dropwise into a solution of the emulsion mixture, or the like method.

The concentration of the emulsion mixture may be determined optionally depending on productivity and is not limited particularly. The concentration is usually from 5 to 50 % by weight, preferably from 10 to 30 % by weight. It is possible that the emulsion mixture is diluted with pure water 2 to 10 times before the coagulation.

Concretely there are co-coagulation methods, for instance, a salting out method, an acid coagulation method, a freeze coagulation method, a method of applying a mechanical shearing force, and the like.

As a coagulating agent, there can be used, for example, an acid such as a nitric acid, hydrochloric acid or sulfuric acid; a metal salt such as aluminum nitrate or aluminum sulfate; or the like. Among them, the acid is preferred from the viewpoint of keeping the polymer clean, and the metal salt is preferred from the viewpoint of easiness of handling.

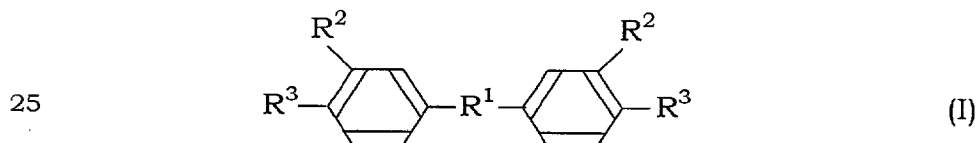
A product obtained by the co-coagulation is washed as the case demands, and dried in a hot air dryer or a vacuum dryer, and thus can be used as a base material for molding.

When the elastomer composition of the present invention is used as a base material, excellent mechanical strength, abrasion resistance, transparency and mold-processability can be imparted to the obtained molded article.

To the elastomer composition of the present invention can be added a crosslinking agent and further a crosslinking accelerator and thus a crosslinkable elastomer composition can be obtained.

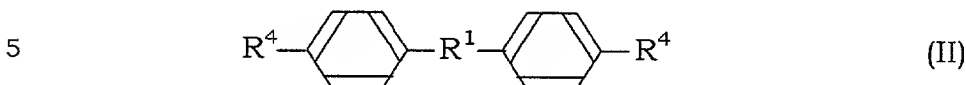
As a crosslinking system, those which have been usually used for an elastomer can be used. Examples thereof are, for instance, an oxazole crosslinking system, imidazole crosslinking system, thiazole crosslinking system, triazine crosslinking system, peroxide crosslinking system, polyol crosslinking system, polyamine crosslinking system, and the like. Also crosslinking with radiant ray, electron beam, ultraviolet ray, or the like can be carried out.

Examples of the crosslinking agent which is used in oxazole crosslinking system, imidazole crosslinking system and thiazole crosslinking system are, for instance, a tetraamine crosslinking agent or bisamino(thio)phenol crosslinking agent represented by the formula (I):



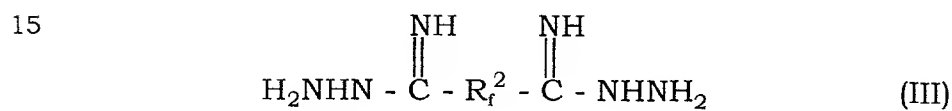
wherein R¹ is -SO₂-, -O-, -CO-, an alkylene group having 1 to 6 carbon atoms, a perfluoroalkylene group having 1 to 10 carbon atoms or a

single bond, one of R^2 and R^3 is $-NH_2$ and another one is $-NH_2$, $-OH$ or $-SH$, a bisamidrazone crosslinking agent represented by the formula (II):

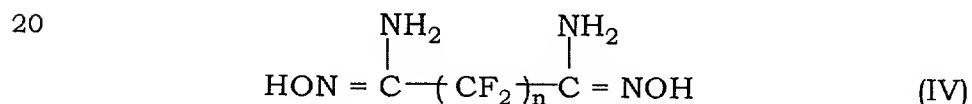


10 (wherein R^1 is as defined above, R^4 is $-\text{C} \begin{matrix} \text{=NH} \\ \text{NHNH}_3 \end{matrix}$ or $-\text{C} \begin{matrix} \text{=NOH} \\ \text{NH}_2 \end{matrix}$),

and a bisamidoxime crosslinking agent represented by the formula (III) or (IV):



wherein R_f^2 is a perfluoroalkylene group having 1 to 10 carbon atoms,



wherein n is an integer of 1 to 10, and the like.

25 As the case demands, a crosslinking accelerator may be used together.

An amount of the crosslinking agent is from 0.5 to 10 parts by weight, preferably from 1 to 5 parts by weight based on 100 parts by weight of the elastomer, and an amount of the crosslinking accelerator is from 0.1 to 10 parts by weight, preferably from 0.2 to 5 parts by weight

based on 100 parts by weight of the elastomer.

Examples of the crosslinking agent to be used in the peroxide crosslinking system are, for instance, 1,1-bis(t-butylperoxy)-3,5,5-trimehtylcyclohexane, 2,5-dimehtylhexane-2,5-dihydroperoxide, di-t-
5 butylperoxide, t-butylcumylperoxide, dicumylperoxide, α,α -bis(t-butylperoxy)-p-diisopropylbenzene, 2,5-dimethyl-2,5-di(t-butylperoxy)hexane, 2,5-dimethyl-2,5-di(t-butylperoxy)-hexyne-3, benzoylperoxide, t-butylperoxybenzene, 2,5-dimethyl-2,5-di(benzoylperoxy)hexane, t-butylperoxymaleate, t-butylperoxyisopropyl
10 carbonate, and the like.

In case of the peroxide crosslinking, it is desirable to use a crosslinking accelerator. Examples of the crosslinking accelerator are, for instance, triallyl cyanurate, triallyl isocyanurate, triallyl formal, triallyl trimellitate, N,N'-m-phenylenebismaleimide, dipropargyl
15 terephthalate, diallyl phthalate, tetraallyl terephthalateamide, triallyl phosphate, and the like.

An amount of the crosslinking agent is from 0.05 to 10 parts by weight, preferably from 1.0 to 5 parts by weight based on 100 parts by weight of the elastomer, and an amount of the crosslinking accelerator is
20 from 0.1 to 10 parts by weight, preferably from 0.5 to 5 parts by weight based on 100 parts by weight of the elastomer.

A transparent elastomeric molded article can be provided in case where, like the peroxide crosslinking system, an inorganic filler, etc. does not need to be used for crosslinking, coloring does not occur
25 through crosslinking and a vulcanized article from a so-called pure rubber formulation exhibits a transparency.

The transparent elastomeric molded article of the present

invention obtained by crosslinking and molding has a transparency of not more than 50 %, preferably not more than 40 %, particularly not more than 30 % in a haze value.

Examples of the crosslinking agent which can be used in the polyol crosslinking system are aromatic polyhydroxy compounds to be usually used. For example, there are 2,2-bis(4-hydroxyphenyl)propane (so-called bisphenol A), 2,2-bis(4-hydroxyphenyl)perfluoropropane (so-called bisphenol AF), resorcin, 1,3,5-trihydroxybenzene, 1,7-dihydroxynaphthalene, 2,7-dihydroxynaphthalene, 1,6-dihydroxynaphthalene, 4,4'-dihydroxydiphenyl, 4,4'-dihydroxystilbene, 2,6-dihydroxyanthracene, hydroquinone, catechol, 2,2-bis(4-hydroxyphenyl)butane (so-called "bisphenol B"), 4,4-bis(4-hydroxyphenyl)valerate, 2,2-bis(4-hydroxyphenyl)tetrafluorochloropropane, 4,4'-dihydroxydiphenylsulfone, 4,4'-dihydroxydiphenylketone, tri(4-hydroxyphenyl)methane, 3,3',5,5'-tetrachlorobisphenol A, 3,3',5,5'-tetrabromobisphenol A, alkali metal salt or alkali earth metal salt thereof, and the like.

Also in the polyol crosslinking system, it is preferable to use a crosslinking accelerator together. Examples thereof are, for instance, ammonium compounds, phosphonium compounds, oxonium compounds, sulfonium compounds, and the like. Particularly, tertiary ammonium salt and tertiary phosphonium salt are preferable.

An amount of the crosslinking agent is from 0.5 to 5 parts by weight based on 100 parts by weight of the elastomer, and an amount of the crosslinking accelerator is from 5 to 400 parts by weight, preferably from 10 to 100 parts by weight based on 100 parts by weight of the

crosslinking agent.

As a crosslinking agent to be used for the polyamine crosslinking, there are polyamine compounds. Examples of the polyamine compound are a primary amine or secondary amine in which
5 two or more basic nitrogen atoms are bonded in a molecule thereof. In many cases, those amines are modified to be formed into a salt so that a reaction becomes mild. Examples thereof are, for instance, alkylenediamines such as ethylenediamine carbamate, hexamethylenediamine carbamate and 4,4'-diaminocyclohexylmethane
10 carbamate which are used comparatively well. Also, there can be used Schiff base such as N,N'-dicinnamylidene-1,6-hexamethylenediamine. In addition the aromatic polyamine compounds having poor basic property can be used preferably in combination with other basic compounds. Examples of the other basic compound are, for instance,
15 diphenylguanidine, di-o-triguanidine, diphenylthiourea, 2-mercaptoimidazoline, and in addition, a compound which has -NH_2 and/or -NH- in its molecule and is used as a crosslinking accelerator for a synthetic rubber, a divalent metal hydroxide, and the like.

An adding amount of the crosslinking agent is from 0.1 to 10
20 parts by weight, preferably from 0.5 to 5 parts by weight based on 100 parts by weight of the elastomer.

Examples of the other additives are a filler (excluding the above-mentioned fluorine-containing resin), a pigment, and the like.

Examples of the filler are, for instance, inorganic fillers such
25 as carbon black (particularly graphitized carbon black), silicon oxide, titanium oxide and alumina; organic fillers such as polyimide, and the like. An adding amount thereof is not more than 100 parts by weight,

preferably from 1 to 50 parts by weight based on 100 parts by weight of the elastomer.

The above-mentioned additives such as a crosslinking agent, crosslinking accelerator and filler may be added at the time of the
5 above-mentioned co-coagulation if possible, or may be mixed after the preparation of the composition comprising the elastomer and the fluorine-containing resin fine particles. The mixing method may be a known kneading method by using a roll, or the like method.

A crosslinked molded article can be produced by kneading
10 and crosslinking the thus obtained crosslinkable elastomer composition in which fluorine-containing resin fine particles are finely dispersed. For the kneading, usual methods, for example, kneading by a roll or kneader can be employed. For the molding, usual compression molding, injection molding, extrusion molding and transfer molding methods can
15 be employed. The molding conditions may be the same as conventional ones.

The obtained crosslinked molded article maintains transparency of the matrix elastomer when a filler is not contained therein. Though there is a case that the transparency is lowered
20 somewhat, a transmissivity of visible light is maintained at 20 % or more of that in case of sole use of the elastomer.

The crosslinked molded article of the present invention is excellent in mechanical strength, post-processability, plasma resistance and gas barrier property irrespective of presence or absence of the filler.
25 Also, since the fluorine-containing resin fine particles finely dispersed are hardly separated from the matrix elastomer, for example, even if the molded article is used as a sealing material for semiconductor

production apparatuses, there is less possibility of generating micro-particles.

The elastomer composition of the present invention is suitably used for molded articles shown in Tables 1 to 3 by making the
5 use of its excellent properties.

TABLE 1

Field of industry	Sub-field of industry	Final product	Equipment
Electrical	Semi-conductor	Semi-conductor production apparatus	CVD equipment
		Liquid crystal panel production apparatus	Dry etching equipment
		Plasma panel production apparatus	Wet etching equipment
			Oxidation and diffusion equipment
			Sputtering equipment
Transportation	Vehicle		Ashing equipment
			Cleaning equipment
			Ion implantation equipment
			Engine and peripheral equipment
			AT
			Fuel line and peripheral equipment
		Aircraft	Fuel line
		Rocket	Fuel line
		Ship	Fuel line
			Fuel line

- continued -

- continued -
Parts
O (square) ring, packing, sealing material, tube, roll, coating, lining, gasket, diaphragm, hose
Gasket, shaft seal, valve stem seal, sealing material, hose
Hose, sealing material
O (square) ring, tube, packing, core material of valve, hose, sealing material, diaphragm
Diaphragm, O (square) ring, valve, tube, packing, hose, sealing material
ditto
ditto
- continued -

- continued -

Chemical	Chemical products	Plant	Production line of chemicals such as pharmaceutical, agricultural chemical, coating and resin
			(Petroleum)
Mechanical	Chemicals	Pharmaceuticals	Plug for chemicals
	Photograph	Developing machine	Film developing machine
			X-ray film developing machine
	Printing	Printing machine	Printing roll
	Coating	Coating facilities	Coating roll
	Analyzer and physical and chemical appliances		
Food	Plant		Food processing line
Metal	Steel making	Steel sheet processing facilities	Steel sheet processing roll

- continued -

- continued -

Lining, valve, packing, roll, hose, diaphragm, O (square) ring, tube, sealing material

Plug for chemicals

Roll

Roll

Roll

Roll

Tube

Lining, valve, packing, roll, hose, diaphragm, O (square) ring, tube, sealing material

Roll

TABLE 2

Field of Industry	Characteristics Required
Electrical	Plasma resistance, acid resistance, alkali resistance, amine resistance, ozone resistance, gas resistance, chemical resistance, cleanliness, heat resistance
Transportation	Heat resistance, amine resistance
	Heat resistance, amine resistance
	Fuel resistance, fuel permeability, heat resistance
	Fuel resistance, fuel permeability, heat resistance
	Fuel resistance, fuel permeability, heat resistance
	Fuel resistance, fuel permeability, heat resistance

- continued -

- 24 -

- 24 -

TABLE 3

Field of industry	Parts
Electrical	O ring and sealing material for gate valve of corresponding production equipment
	O ring and sealing material for quartz window of corresponding production equipment
	O ring and sealing material for chamber of corresponding production equipment
	O ring and sealing material for gate of corresponding production equipment
	O ring and sealing material for bell jar of corresponding production equipment
	O ring and sealing material for coupling of corresponding production equipment
	O ring and sealing material for pump of corresponding production equipment
	O ring and sealing material for gas controller for semi-conductor of corresponding production equipment
	O ring and sealing material for resist developing and releasing solutions
	O ring and sealing material for wafer cleaning solution
	Diaphragm of pump for corresponding production equipment
	Hose for resist developing and releasing solutions
	Hose and tube for wafer cleaning solution
	Roll for transferring wafer
	Lining and coating of tanks for resist developing and releasing solutions
	Lining and coating of tanks for wafer cleaning solution
	Lining and coating of tanks for wet etching

- continued -

- continued -

Transportation

Engine head gasket

Metal gasket

Crank shaft seal

Cam shaft seal

Valve stem seal

Manifold packing

Oil hose

ATF hose

Injector O ring

Injector packing

O ring and diaphragm for fuel pump

Fuel hose

- continued -

- continued -

Chemical

Mechanical

Developing roll

Developing roll

Gravure roll

Guide roll

Gravure roll for magnetic tape production and coating line

Guide roll for magnetic tape production and coating line

Various coating rolls

Food

Metal

Particularly the molded articles of the present invention can be used built-in the following semiconductor manufacturing equipment.

(1) Etching system

Dry etching equipment

- 5 Plasma etching machine
- Reactive ion etching machine
- Reactive ion beam etching machine
- Sputter etching machine
- 10 Ion beam etching machine

Wet etching equipment

Ashing equipment

(2) Cleaning system

Dry etching cleaning equipment

- 15 UV/O₃ cleaning machine
- Ion beam cleaning machine
- Laser beam cleaning machine
- Plasma cleaning machine
- Gas etching cleaning machine

Extractive cleaning equipment

- 20 Soxhlet extractive cleaning machine
- High temperature high pressure extractive cleaning machine
- Microwave extractive cleaning machine
- Supercritical extractive cleaning machine

(3) Exposing system

- 25 Stepper
- Coater and developer

(4) Polishing system

CMP equipment

(5) Film forming system

CVD equipment

Sputtering equipment

5 (6) Diffusion and ion implantation system

Oxidation and diffusion equipment

Ion implantation equipment

The present invention is then explained by means of examples and preparation examples, but is not limited to them.

10

PREPARATION EXAMPLE 1

(Preparation of emulsion of fluorine-containing elastomer particles)

A 47-liter stainless steel autoclave having no ignition source was charged with 30 liter of pure water, 300 g of $C_7F_{15}COONH_4$ as an emulsifying agent and 2.7 g of disodium phosphate·12H₂O as a pH control agent. After the inside of a system was sufficiently replaced with nitrogen gas and deairing was carried out, the autoclave was heated to 50°C with stirring at 200 rpm, and a gas mixture of tetrafluoroethylene (TFE)/perfluoro(methyl vinyl ether) (PMVE) (TFE/PMVE=24/76 in mole ratio) was introduced so that the inside pressure became 1.18 MPa. Then 100 ml of an aqueous solution of ammonium persulfate (APS) having a concentration of 55.8 mg/ml was introduced with pressurized nitrogen gas to initiate a reaction.

With the advance of the polymerization, at the time when the inside pressure was lowered to 1.08 MPa, 62.3 g of di-iodine compound $[I(CF_2)_4I]$ was introduced with pressurized nitrogen gas. Then 60 g of TFE was introduced at its self-pressure and 60 g of PMVE was

introduced with a plunger pump so that the inside pressure became 1.18 MPa. Thereafter with the advance of the reaction, pressurized TFE and PMVE were introduced similarly. Thus increasing and decreasing of the inside pressure were repeated between 1.08 MPa and 1.18 MPa, and at the time when a total amount of TFE and PMVE reached 6.5 kg, 7.8 kg, 9.1 kg and 10.4 kg, respectively, 25.6 g of an iodine compound $\text{CF}_2=\text{CFOCF}_2\text{CF}_2\text{CH}_2\text{I}$ was introduced with pressurized nitrogen gas. Every 12 hours after starting of the polymerization, 20 ml of an aqueous solution of APS having a concentration of 52.5 mg/ml was introduced with pressurized nitrogen gas.

When a total amount of the introduced TFE and PMVE reached 13 kg 33 hours after starting of the polymerization reaction, the autoclave was cooled and an un-reacted monomer was released to obtain an emulsion (A-1) of fluorine-containing elastomer particles (average particle size: 70 nm) having a solid concentration of 27.5 % by weight.

A part of the emulsion was sampled and nitric acid was added thereto for coagulation. The coagulated product was washed and dried to obtain elastomer particles. The Mooney viscosity ML_{1+10} (100°C) of the elastomer was 42, and according to ^{19}F -NMR analysis, the components thereof was TFE/PMVE=62/38 (% by mole). The glass transition temperature T_g (center value) measured according to DSC was -3°C.

25

PREPARATION EXAMPLE 2

(Preparation of emulsion of fluorine-containing resin fine particles)

A 6-liter stainless steel autoclave having no ignition source

was charged with 3 liter of pure water, 30 g of $C_3F_7OCF(CF_3)CF_2OCF(CF_3)COONH_4$ as an emulsifying agent and 0.27 g of disodium phosphate·12H₂O as a pH control agent. After the inside of a system was sufficiently replaced with nitrogen gas and deairing was carried out, the autoclave was heated to 80°C with stirring at 600 rpm, and a gas mixture of tetrafluoroethylene (TFE)/perfluoro(methyl vinyl ether) (PMVE) (TFE/PMVE=88/12 in mole ratio) was introduced so that the inside pressure became 0.20 MPa. Then 4 ml of an aqueous solution of ammonium persulfate (APS) having a concentration of 2.5 mg/ml was introduced with pressurized nitrogen gas to initiate a reaction.

With the advance of the polymerization, at the time when the inside pressure was lowered to 0.15 MPa, a gas mixture of TFE/PMVE (TFE/PMVE=95/5 in mole ratio) was introduced with pressurized nitrogen gas so that the inside pressure became 0.20 MPa. Thereafter with the advance of the reaction, pressurized TFE and PMVE gas mixture (95/5 in mole ratio) was introduced similarly. Thus increasing and decreasing of the inside pressure were repeated between 0.15 MPa and 0.20 MPa.

When a total amount of the introduced TFE and PMVE reached 331 g 4.5 hours after starting of the polymerization reaction, the autoclave was cooled and an un-reacted monomer was released to obtain an emulsion (B-1) of fluorine-containing resin fine particles (average particle size: 44 nm) having a solid concentration of 9.7 % by weight. The average particle size was measured by mixing 120 mg of the emulsion and 4.4 g of dimethylsulfoxide and measuring with LPA-3000, 3100 available from OTSUKA DENSHI KABUSHIKI KAISHA.

A part of the emulsion was sampled and nitric acid was added thereto for coagulation. The coagulated product was washed and dried to obtain a white fluorine-containing resin fine powder. The melt flow rate MFR of the fluorine-containing resin was un-measurable under conditions of maintaining at 372°C for five minutes. According to ^{19}F -NMR analysis, the components thereof was TFE/PMVE=94.5/5.5 (% by mole). The melting point measured according to DSC was 290°C.

PREPARATION EXAMPLE 3

(Preparation of emulsion of fluorine-containing resin fine particles)

A 6-liter stainless steel autoclave having no ignition source was charged with 3 liter of pure water, 30 g of $\text{C}_3\text{F}_7\text{OCF}(\text{CF}_3)\text{CF}_2\text{OCF}(\text{CF}_3)\text{COONH}_4$ as an emulsifying agent and 0.27 g of disodium phosphate $\cdot 12\text{H}_2\text{O}$ as a pH control agent. After the inside of a system was sufficiently replaced with nitrogen gas and deairing was carried out, the autoclave was heated to 80°C with stirring at 600 rpm, and a gas mixture of tetrafluoroethylene (TFE)/perfluoro(methyl vinyl ether) (PMVE) (TFE/PMVE=88/12 in mole ratio) was introduced so that the inside pressure became 0.20 MPa. Then 4 ml of an aqueous solution of ammonium persulfate (APS) having a concentration of 2.5 mg/ml was introduced with pressurized nitrogen gas to initiate a reaction.

With the advance of the polymerization, at the time when the inside pressure was lowered to 0.15 MPa, 2.92 g of a di-iodine compound $[\text{I}(\text{CF}_2)_4\text{I}]$ was introduced with pressurized nitrogen gas and then a gas mixture of TFE/PMVE (TFE/PMVE=95/5 in mole ratio) was introduced with pressurized nitrogen gas so that the inside pressure became 0.20

MPa. Thereafter with the advance of the reaction, pressurized TFE/PMVE gas mixture (95/5 in mole ratio) was introduced similarly. Thus increasing and decreasing of the inside pressure were repeated between 0.15 MPa and 0.20 MPa.

5 When a total amount of the introduced TFE and PMVE
reached 327 g 9.9 hours after starting of the polymerization reaction, the
autoclave was cooled and an un-reacted monomer was released to
obtain an emulsion (B-2) of fluorine-containing resin fine particles
(average particle size: 44 nm) having a solid concentration of 9.4 % by
10 weight.

A part of the emulsion was sampled and nitric acid was added thereto for coagulation. The coagulated product was washed and dried to obtain a white fluorine-containing resin fine powder. The melt flow rate MFR of the fluorine-containing resin was 21.1 g/10 min under conditions of maintaining at 372°C for five minutes. According to ^{19}F -NMR analysis, the components thereof was TFE/PMVE=94.9/5.1 (% by mole). The melting point measured according to DSC was 280.7°C.

20 PREPARATION EXAMPLE 4

(Preparation of emulsion of fluorine-containing elastomer particles)

A 3-liter stainless steel autoclave having no ignition source was charged with 1 liter of pure water, 10 g of $C_3F_7OCF(CF_3)CF_2OCF(CF_3)COONH_4$ as an emulsifying agent and 0.09 g of disodium phosphate·12H₂O as a pH control agent. After the inside of a system was sufficiently replaced with nitrogen gas and deairing was carried out, the autoclave was heated to 53°C with stirring at 600 rpm,

and a gas mixture of tetrafluoroethylene (TFE)/perfluoro(methyl vinyl ether) (PMVE) (TFE/PMVE=25/75 in mole ratio) was introduced so that the inside pressure became 0.78 MPa. Then 20 ml of an aqueous solution of ammonium persulfate (APS) having a concentration of 264
5 mg/ml was introduced with pressurized nitrogen gas to initiate a reaction.

With the advance of the polymerization, at the time when the inside pressure was lowered to 0.69 MPa, 2.2 g of $\text{CF}_2=\text{CFOCF}_2\text{CF}(\text{CF}_3)\text{OCF}_2\text{CF}_2\text{CN}$ (CNVE) was introduced with
10 pressurized nitrogen gas. Then 4.7 g of TFE and 5.3 g of PMVE were introduced at the respective self-pressures. Thereafter with the advance of the reaction, pressurized TFE and PMVE were introduced similarly. Thus increasing and decreasing of the inside pressure were repeated between 0.69 MPa and 0.78 MPa, and in addition, at the time
15 when a total amount of the introduced TFE and PMVE reached 70 g, 2.2 g of CNVE was introduced with pressurized nitrogen gas.

When a total amount of the introduced TFE and PMVE reached 130 g six hours after starting of the polymerization reaction, the autoclave was cooled and an un-reacted monomer was released to
20 obtain 1,160 g of emulsion (A-2) of fluorine-containing elastomer particles having a solid concentration of 11.3 % by weight.

Then 100 g of the obtained emulsion was diluted with 300 g of water and added slowly to 280 g of an aqueous solution of 3.5 % by weight of hydrochloric acid with stirring. After the addition, stirring
25 was carried out for five minutes and a coagulated product was filtrated. The obtained elastomer particles were further washed with 200 g of HCFC-141b and filtrated. After the washing with HCFC-141b and the

filtration were repeated four times, vacuum drying was carried out at 60°C for 72 hours to obtain 11.2 g of fluorine-containing elastomer.

As a result of ^{19}F -NMR analysis, the obtained fluorine-containing elastomer was one comprising TFE/PMVE/CNVE =
5 60.4/38.9/0.7 (% by mole).

EXAMPLE 1

The emulsion of the fluorine-containing elastomer particles (A-1) prepared in Preparation Example 1 and the emulsion of the
10 fluorine-containing resin fine particles (B-1) prepared in Preparation Example 2 were mixed in the amounts of 1,236 g and 619 g, respectively (solid content ratio: fluorine-containing elastomer/fluorine-containing resin = 85/15 in weight ratio), and the mixture was added dropwise over
15 10 minutes to 411 g of a 9 % aqueous solution of nitric acid being stirred and thus subjected to co-coagulation. The obtained co-coagulate was washed with water and dried to obtain an elastomer composition comprising the fluorine-containing resin fine particles finely dispersed
in the fluorine-containing elastomer.

When the elastomer composition was subjected to DTA
20 measurement, there was recognized at 272.6°C an absorption considered to be derived from the fluorine-containing resin. The composition was transparent (haze value: 12 %).

(Measurement of haze value)

A 0.7 mm thick sample is produced from the elastomer
25 composition at a melting temperature thereof (60° to 150°C) by applying a pressure thereto if necessary. A haze value of the sample is measured with a direct current haze meter (a measuring device according to JIS

K7105 and ASTM D 1003 available from TOYO SEIKI KABUSHIKI KAISHA).

A haze value of a crosslinked molded article described hereinbelow is measured by using a 2 mm thick sample sheet produced
5 by crosslinking and molding the elastomer composition.

Fig. 1 shows a photograph of the elastomer composition taken at $\times 20000$ magnification with a transmission type electron microscope (TEM) by a replica method. In Fig. 1, convex portions in the form of particle are fine particles of the fluorine-containing resin. It can
10 be seen that many fluorine-containing resin fine particles (average particle size: about 40 nm) are finely dispersed uniformly. This finely dispersed state clearly differs from that of a composition in a TEM photograph (Fig. 2) obtained in Comparative Example 3 described hereinbelow.

15

EXAMPLE 2

Co-coagulation was carried out in the same manner as in Example 1 except that a mixing amount of the emulsion (B-1) of the fluorine-containing resin fine particles was changed to 412 g, to obtain
20 an elastomer composition (solid content ratio: fluorine-containing elastomer/fluorine-containing resin = 90/10 in weight ratio). The composition was transparent (haze value: 12 %).

EXAMPLE 3

25 Co-coagulation was carried out in the same manner as in Example 1 except that a mixing amount of the emulsion (B-1) of the fluorine-containing resin fine particles was changed to 206 g, to obtain

an elastomer composition (solid content ratio: fluorine-containing elastomer/fluorine-containing resin = 95/5 in weight ratio). The composition was transparent (haze value: 11 %).

5

EXAMPLE 4

Co-coagulation was carried out in the same manner as in Example 1 except that 638 g of the emulsion (B-2) of the fluorine-containing resin fine particles obtained in Preparation Example 3 was mixed instead of the emulsion (B-1) of the fluorine-containing resin fine
10 particles, to obtain an elastomer composition (solid content ratio: fluorine-containing elastomer/fluorine-containing resin = 85/15 in weight ratio). The composition was transparent (haze value: 12 %).

EXAMPLE 5

15 To 100 parts by weight of the elastomer composition prepared in Example 1 were mixed 1 part by weight of 2,5-dimethyl-2,5-di(t-butylperoxy)hexane as a crosslinking agent and 1 part by weight of triallyl isocyanurate as a crosslinking accelerator. The mixture was kneaded with an open roll to obtain a crosslinkable elastomer
20 composition.

Vulcanizability of the crosslinkable elastomer composition was determined by a method described hereinbelow. The results are shown in Table 4.

Further the crosslinkable elastomer composition was
25 subjected to press-crosslinking at 150°C for 30 minutes and then crosslinking in an oven at 180°C for four hours to obtain a crosslinked article. Physical properties in normal state of the crosslinked article

were measured. Also an O-ring (P-24) was produced under the same crosslinking conditions and a compression set thereof was measured. Further a haze value of the crosslinked elastomer sheet (2 mm thick) was measured. The results are shown in Table 4.

5 (Vulcanizability)

A vulcanization curve of each composition for vulcanization is obtained at a temperature shown in Table 4 by using a JSR Curastometer Model II, and a minimum torque, maximum torque, induction time and optimum vulcanization time are obtained therefrom.

10 (Physical properties in normal state)

100 % modulus, tensile strength, tensile elongation and hardness (JIS A hardness) in normal state (25°C) are measured according to JIS K6301.

(Compression set)

15 A compression set after allowing to stand at 200°C for 70 hours is measured according to JIS K6301.

EXAMPLES 6 to 10

20 A crosslinkable elastomer composition having components shown in Table 4 was prepared in the same manner as in Example 5 and vulcanizability thereof was determined. Also a crosslinked article was produced under the same crosslinking conditions as in Example 5, and physical properties in normal state, compression set and haze value were determined. The results are shown in Table 4.

25 A filler mixed in Examples 6 to 8 is a silicon oxide (AEROSIL 300 available from NIPPON AEROSIL KABUSHIKI KAISHA).

COMPARATIVE EXAMPLES 1 to 2

A crosslinkable elastomer composition (haze value: 10 %) was prepared in the same manner as in Example 5 except that fluorine-containing resin fine powder was not mixed. Vulcanizability of the composition was determined, and also a crosslinked article was produced under the same crosslinking conditions as in Example 5, and physical properties in normal state and compression set were determined. The results are shown in Table 4.

COMPARATIVE EXAMPLE 3

To the emulsion (A-1) of the fluorine-containing elastomer particles obtained in Preparation Example 1 was added a nitric acid for coagulation thereof, and the coagulate was washed and dried to obtain elastomer particles. On the other hand, to the emulsion (B-1) of the fluorine-containing resin fine particles obtained in Preparation Example 2 was added a nitric acid for coagulation thereof, and the coagulate was washed and dried to obtain a white fluorine-containing resin powder. To 85 parts by weight of the elastomer particles was dry-blended 15 parts by weight of the fluorine-containing resin powder to obtain a white opaque composition (haze value: 82 %). A TEM photograph (×20000) of the composition is shown in Fig. 2. As it is clear from Fig. 2, an elastomer portion (flat portions at both sides in Fig. 2) is separated clearly from a portion where fluorine-containing resin fine particles are agglomerated (a portion having a coarse surface at a center in Fig. 2).

A crosslinkable elastomer composition was prepared in the same manner as in Example 5 except that the above-mentioned composition was used, and vulcanizability thereof was determined.

Also a crosslinked article was produced under the same crosslinking conditions as in Example 5, and physical properties in normal state, compression set and haze value were determined. The results are shown in Table 4.

TABLE 4

Elastomer composition	Ex. 5	Ex. 6	Ex. 7	Ex. 8	Ex. 9	Ex. 10	Com. Ex. 1	Com. Ex. 2	Com. Ex. 3
Example 1	100	100				100			
Example 2			100						
Example 3				100					
Example 4					100				
Preparation Example 1 (elastomer)							100	100	85
Preparation Example 2 (fluorine- containing resin powder)									15
Crosslinking agent	1	1	1	1	1	1	1	1	1
Crosslinking accelerator	1	3	3	3	1	0.5	1	3	1
SiO ₂	-	3	3	3	-	-	-	3	-

- continued -

- continued -

Vulcanizability (160°)										
Minimum torque (kg)	0.18	0.40	0.27	0.15	0.10	0.17	0.03	0.10	0.06	
Maximum torque (kg)	4.51	7.70	7.20	6.15	5.02	4.12	3.98	6.00	4.71	
Induction time (min)	0.7	0.5	0.5	0.5	0.6	0.7	0.5	0.6	0.5	
Optimum vulcanization time (min)	1.4	1.4	1.2	1.4	1.5	1.4	1.1	1.1	0.9	
Physical properties in normal state										
100 % modulus (MPa)	4.5	13.8	7.4	6.4	3.7	4.5	2.1	4.9	4.7	
Tensile strength (MPa)	20.8	24.9	22.8	16.3	18.3	19.7	12.3	16.5	8.6	
Elongation (%)	202	155	208	152	197	208	186	180	168	
Hardness (JIS A)	71	80	75	71	71	70	60	68	71	
Compression set (%)	33	27	22	12	34	36	18	13	37	
Haze value (%)	41	-	-	-	43	20	-	-	82	

EXAMPLE 11

The fluorine-containing elastomer particles (A-2) prepared in Preparation Example 4 and the fluorine-containing resin fine particles (B-1) prepared in Preparation Example 2 were mixed in the amounts of 300 g and 619 g, respectively in a ratio of fluorine-containing elastomer/fluorine-containing resin = 85/15 (weight ratio), and the mixture was added dropwise over 20 minutes for co-coagulation to 981 g of a 9 % aqueous solution of nitric acid being stirred. The obtained coagulate was washed with water and dried to obtain a transparent elastomer composition (haze value: 18 %) comprising the fluorine-containing resin fine particles finely dispersed in the fluorine-containing elastomer.

EXAMPLE 12

A crosslinkable elastomer composition was prepared by kneading 1.45 parts by weight of 2,2bis-[(3-amino-4-phenylamino)phenyl]hexafluoropropane (synthesized by a method described in Journal of Polymer Science, edited by Polymer Chemistry, Vol. 20, 2381 to 2393 (1982)) as a crosslinking agent based on 100 parts by weight of the elastomer composition prepared in Example 11 by using an open roll. Vulcanizability of the obtained crosslinkable elastomer composition which was determined by the above-mentioned method was as follows.

Vulcanizability (170°C)

Minimum torque: 0.45 kg
Maximum torque: 2.90 kg
Induction time: 4.4 min

Optimum vulcanization time: 8.7 min

Further the crosslinkable elastomer composition was subjected to press-crosslinking at 170°C for 15 minutes and crosslinking in an oven at 204°C for 18 hours and then at 288°C for 18
5 hours to obtain a crosslinked article. Physical properties in normal state of the obtained crosslinked article was also determined in the same manner as above, and also an O-ring (P-24) was produced under the same conditions as above to determine its compression set. The results are as follows.

10 Physical properties in normal state

100 % modulus: 2.9 MPa

Tensile strength: 19.4 MPa

Elongation: 252 %

Hardness (JIS A): 74

15 Compression set (200°C, 70 hours, 25 % compression)

Compression set: 15 %

INDUSTRIAL APPLICABILITY

The present invention can provide a composition capable of
20 giving an elastomeric molded article excellent in mechanical strength, abrasion resistance, transparency, etc.

CLAIMS

1. A transparent elastomer composition comprising an elastomer and fluorine-containing resin fine particles which are finely dispersed in the elastomer.
5

2. A composition comprising a transparent elastomer composition which comprises an elastomer and fluorine-containing resin fine particles which are finely dispersed in the elastomer and further containing a filler.
10

3. The composition of Claim 1 or 2, wherein the elastomer is a fluorine-containing elastomer.

4. The composition of any of Claims 1 to 3, wherein the composition contains a crosslinking agent.
15

5. The composition of any of Claims 1 to 4, wherein the transparent elastomer composition has a haze value of not more than 50 %.
20

6. A process for preparing the transparent elastomer composition of Claim 1, which comprises mixing an emulsion of elastomer particles and an emulsion of fluorine-containing resin fine particles, and co-coagulating the mixture.
25

7. The process of Claim 6, wherein the emulsion of fluorine-containing resin fine particles is an emulsion of fluorine-containing resin fine particles having an average particle size of from 20 to 150 nm.

5 8. An elastomeric molded article which is obtained by vulcanizing and molding the composition of any of Claims 1 to 5.

9. The elastomeric molded article of Claim 8, which is transparent.

10

10. The elastomeric molded article of Claim 9, which has a haze value of not more than 50 %.

ABSTRACT

A transparent elastomer composition obtained by coagulation of an emulsion of fluorine-containing resin fine particles
5 having an average particle size of from 20 to 150 nm and an emulsion of elastomer particles, in which the fluorine-containing resin fine particles are finely dispersed uniformly in the elastomer is obtained. The transparent elastomer composition provides an elastomeric molded article having excellent mechanical strength, abrasion resistance and
10 transparency.

FIG. 1



1 μ m

FIG. 2



1 μ m

DECLARATION AND POWER OF ATTORNEY FOR PATENT APPLICATION
特許出願宣言書
Japanese Language Declaration

私は、下欄に氏名を記載した発
明者として、以下のとおり宣言
する：

私の住所、郵便の宛先および国
籍は、下欄に氏名に続いて記載
したとおりであり、

名称の発明に関し、請求の範囲
に記載した特許を求める主題の
本来の、最初にして唯一の発明
者である(一人の氏名のみが下欄
に記載されている場合)か、もし
くは本来の、最初にして共同の
発明者である(複数の氏名が下欄
に記載されている場合)と信じ、

As a below-named inventor, I
hereby declare that:

My residence, post office
address and citizenship are as
stated below next to my name,

I believe I am the original,
first and sole inventor (if
only one name is listed below)
or an original, first and
joint inventor (if plural
names are listed below) of the
subject matter which is
claimed and for which a patent
is sought on the invention
entitled:

TRANSPARENT ELASTOMER COMPOSITION

その明細所を
(該当する方に印を付す)

☐ ここに添付する。

☐ 第 _____ 日に出席番号第
_____ 号として提出し、
_____ 日に補正した。
(該当する場合)

私は、前記のとおり補正した請
求の範囲を含む前記明細書の内
容を検討し、理解したことを陳
述する。

私は、連邦規則法典第37部第1章
第56条(a)項に従い、本願の審査
に所要の情報を開示すべき義務
を有することを認める。

the specification of which
(check one)

☐ is attached hereto

☒ was filed on September 27, 2000 as
PCT International
Appln. Ser. No. PCT/JF00/06631
and was amended on _____
(if applicable)

I hereby state that I have
reviewed and understood the
contents of the above-
identified specification,
including the claims, as
amended by any amendments
referred to above.

I acknowledge the duty to
disclose information in
accordance with Title 37, Code
of Federal Regulations, §
1.56(a).

私には、合衆国法典第35部第119条
 にも、とつく下記の外国の特許出願の
 ままた、は、明者証の外国の特許出願の
 主張を主張する者証を明者証する
 前出願の明記する

I hereby claim foreign
 priority benefits under Title
 35, United States Code, § 119
 of any foreign application(s)
 for patent or inventor's cer-
 tificate having a filing date
 before that of the application
 on which priority is claimed:

Prior foreign applications:
 先の外国出願:

Priority claimed
 優先権の主張

277995/1999 Japan
 Number Country
 番号 国名

September 30, 1999
 Filing date
 出願の年月日

☒ ☐
 Yes No
 あり なし

Number Country
 番号 国名

Filing date
 出願の年月日

☐ ☐
 Yes No
 あり なし

Number Country
 番号 国名

Filing date
 出願の年月日

☐ ☐
 Yes No
 あり なし

私には、合衆国法典第35部第120条
 のも、とつく下記の外国の特許出願の
 範囲の各主記の合衆国の主記の請求
 定法典第35部第112条第1項に開
 示の態様で先行願の合衆国と本願の
 先出願の間に公表されたPCT条(a)
 の第37部の第1章第56条(a)項に
 の所有する情報を開示することを認める:

I hereby claim the benefit
 under Title 35, United States
 Code, § 120 of any United
 States application(s) listed
 below and, insofar as the
 subject matter of each of the
 claims is not disclosed in the
 prior United States applica-
 tion in the manner provided by
 the first paragraph of Title
 35, United States Code, § 112,
 I acknowledge the duty to
 disclose material information
 as defined in Title 37, Code
 of Federal Regulations, §
 1.56(a) which occurred between
 the filing date of the prior
 of the prior application and
 the national or PCT
 International filing date of
 this application:

Appln. Ser. No.
 出願番号

Filing Date
 出願日

Status (patd., pend., abndnd.)
 現況(特許済み、係属中、放棄済み)

Appln. Ser. No.
 出願番号

Filing Date
 出願日

Status (patd., pend., abndnd.)
 現況(特許済み、係属中、放棄済み)

私はい、こ、こ、に、自、己、の、知、識、に、も、と
づ、て、行、こ、っ、に、陳、述、の、有、す、る、情、報、を、真、実、と
び、信、ず、る、と、こ、で、偽、り、の、陳、述、を、行、つ、た、ら、ば、第、一、に、罰、金、を、課、せ、ら、れ、る、こ、と、に、
陳、述、に、お、き、よ、り、科、せ、ら、れ、る、罰、金、を、課、せ、ら、れ、る、こ、と、に、
場、合、に、お、き、よ、り、科、せ、ら、れ、る、罰、金、を、課、せ、ら、れ、る、こ、と、に、
条、に、お、き、よ、り、科、せ、ら、れ、る、罰、金、を、課、せ、ら、れ、る、こ、と、に、
処、に、お、き、よ、り、科、せ、ら、れ、る、罰、金、を、課、せ、ら、れ、る、こ、と、に、
刑、に、お、き、よ、り、科、せ、ら、れ、る、罰、金、を、課、せ、ら、れ、る、こ、と、に、
に、お、き、よ、り、科、せ、ら、れ、る、罰、金、を、課、せ、ら、れ、る、こ、と、に、
本、に、お、き、よ、り、科、せ、ら、れ、る、罰、金、を、課、せ、ら、れ、る、こ、と、に、
有、に、お、き、よ、り、科、せ、ら、れ、る、罰、金、を、課、せ、ら、れ、る、こ、と、に、
効、に、お、き、よ、り、科、せ、ら、れ、る、罰、金、を、課、せ、ら、れ、る、こ、と、に、
認、に、お、き、よ、り、科、せ、ら、れ、る、罰、金、を、課、せ、ら、れ、る、こ、と、に、
を、に、お、き、よ、り、科、せ、ら、れ、る、罰、金、を、課、せ、ら、れ、る、こ、と、に、
た、に、お、き、よ、り、科、せ、ら、れ、る、罰、金、を、課、せ、ら、れ、る、こ、と、に、

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

委任状：私は、下記発明者として、以下の代理人をここに選任し、本願の権利を遂行すること並びにこれに關する一切の行為を特許商標庁に対して行うことを委任する。

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorneys to prosecute this application and transact all business in the Patent and Trademark Office connected therewith.

I hereby appoint John H. Mion, Reg. No. 18,879; Donald E. Zinn, Reg. No. 19,046; Thomas J. Macpeak, Reg. No. 19,292; Robert Seas, Jr., Reg. No. 21,092; Darryl Mexic, Reg. No. 23,063; Robert V. Sloan, Reg. No. 22,775; Peter D. Olexy, Reg. No. 24,513; J. Frank Osha, Reg. No. 24,625; Waddell A. Biggart, Reg. No. 24,861; Robert G. McMorrow, Reg. No. 19,093; Louis Gubinsky, Reg. No. 24,835; Neil B. Siegel, Reg. No. 25,200; David J. Cushing, Reg. No. 28,703; John R. Inge, Reg. No. 26,916; Joseph J. Ruch, Jr., Reg. No. 26,577; Sheldon I. Landsman, Reg. No. 25,430; Richard C. Turner, Reg. No. 29,710; Howard L. Bernstein, Reg. No. 25,665; Alan J. Kasper, Reg. No. 25,426; Kenneth J. Burchfiel, Reg. No. 31,333; Gordon Kit, Reg. No. 30,764; Susan J. Mack, Reg. No. 30,951; Frank L. Bernstein, Reg. No. 31,484; Mark Boland, Reg. No. 32,197; William H. Mandir, Reg. No. 32,156; Scott M. Daniels, Reg. No. 32,562; Brian W. Hannon, Reg. No. 32,778; Abraham J. Rosner, Reg. No. 33,276; Bruce E. Kramer, Reg. No. 33,725; Paul F. Neils, Reg. No. 33,102; and Brett S. Sylvester, Reg. No. 32,765, my attorneys to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith, and request that all correspondence about the application be addressed to SUGHRUE, MION, ZINN, MACPEAK & SEAS, 2100 Pennsylvania Avenue, N.W., Washington, D.C. 20037-3202.

Full name of sole or first inventor

唯一のまたは第一の発明者の氏名(α-マシ*)

Sadashige IRIE

Inventor's signature

Date

同発明者の署名

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Fourth Inventor's signature

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Full name of sixth joint inventor (if any)

第6の共同発明者の氏名(該当する場合)(ロ-マシ*) Mitsuru KISHINE

Sixth Inventor's signature

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